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Observation of Atomic Relaxation Near an Interface
Through Detection of Emitted Fluorescence

by

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OBSERVATION OF ATOMIC RELAXATION NEAR AN INTERFACE THROUGH DETECTION
OF EMITTED FLUORESCENCE

ABSTRACT

In this Comment we discuss the general features of spontaneous decay of an atom which is close to the surface of a dielectric (or metal) layer, and this is compared to atomic decay near a four-wave mixing phase conjugator. It is pointed out that the decay or relaxation constants can be expressed entirely in terms of the classical Fresnel coefficients for reflection and transmission of a plane wave, independent of what the explicit forms of these coefficients are. For decay near a dielectric the relaxation constants can be measured directly by counting the number of fluorescent photons per unit of time, after excitation by a laser. For relaxation near a phase conjugator this procedure would require a spectral resolution in the photon detection, since there are two distinct contributions to the fluorescent yield of a two-state atom. The two possible decay mechanisms have different relaxation constants and produce radiation with different frequencies.

Keywords: Relaxation, Fluorescence, Lifetimes, Phase Conjugation

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When an atom is in the vicinity of an optically-reflecting surface, it behaves differently than in empty space. For instance, the mechanism of emission of fluorescence radiation is affected by the presence of the medium, and also the details of the dynamics of spontaneous decay from an excited atomic state are different. We consider a single atom in vacuum which is separated by a normal distance h from a planar substrate, such that h is on the order of an optical wavelength. Since h is much larger than the dimensions of an atom, the electronic wave functions of the atom do not extend into the region of the substrate material, and hence the atom can be considered to be surrounded by vacuum only. Nevertheless, the rate of spontaneous atomic decay is altered by the medium, as was recently demonstrated experimentally for atoms near a silver mirror,^{1,2} which leads to the inevitable conclusion that the optical vacuum near an interface is different from the vacuum of completely empty space. In this Comment we discuss the rather general situation where the medium is an isotropic dielectric layer (DL), which can be specified by its dielectric constant ϵ and layer thickness. The limit $\epsilon \rightarrow -\infty$ then corresponds to a perfectly-conducting substrate (mirror), which pertains to the quoted experiments. In addition, we shall look at the more exotic situation where the medium is a transparent four-wave mixer, e.g., a nonlinear crystal which is pumped by two strong counterpropagating laser beams. Such a device effectively acts as a phase conjugator^{3,4} (PC), and we shall show that the modified vacuum near a PC gives rise to atomic behavior which can never occur near any linear medium.

Spontaneous decay is a relaxation phenomenon, which is most conveniently described with reservoir theory.^{5,6} For a model two-state atom this leads in general to the set of equations

$$\frac{dn_e}{dt} = -A_e n_e + A_g n_g$$

$$\frac{dn_g}{dt} = -A_g n_g + A_e n_e$$

for the populations n_e and n_g of the excited state and ground state, respectively. When other relaxation or excitation mechanisms or other decay channels are present, then additional terms appear on the right-hand sides of the two equations. The explicit form of the relaxation constants A_e and A_g is determined by the details of the mechanism which is responsible for the relaxation, and they can be evaluated with reservoir theory. The set of equations has the form of a set of rate (or gain-loss) equations, from which it follows that $A_e n_e$ equals the number of transitions per unit time from the excited state to the ground state, and a similar interpretation holds for the term $A_g n_g$. For spontaneous decay in empty space or near a DL, we must obviously have $A_g = 0$, since the term $A_g n_g$ would correspond to a spontaneous excitation of the atom, which is prohibited by energy conservation. Consequently, the only relaxation constant is A_e , and the solution for n_e is readily found to be $n_e(t) = n_e(0)\exp(-A_e t)$, which is the usual exponential decay. The parameter A_e is the Einstein A-coefficient, which can be measured experimentally by observing the number of excited atoms as a function of time, after an excitation with, for instance, a laser. It turns out that the value of A_e for an atom near a medium is different than for an atom in empty space, as was shown experimentally^{1,2} and predicted theoretically.⁷ Both an enhancement and a decrease of the decay rate can occur, as illustrated in Fig. 1 for the case of a mirror.

The question can be raised as to what the reason is for this change in lifetime ($1/A_e$) of an excited atomic level when the atom is close to a boundary with a dielectric medium. First we recall that for an atom in empty space the mechanism of spontaneous decay is brought about by a coupling between the atomic dipole moment operator $\underline{\mu}$ and the electric component \underline{E} of the vacuum radiation field, according to the interaction Hamiltonian $H_I = -\underline{\mu} \cdot \underline{E}$. Here \underline{E} has to be evaluated at the position of the atom. The explicit form of \underline{E} is the standard expansion into traveling plane waves, which have the photon creation and annihilation operators as amplitude factors.⁸ For a description of spontaneous decay it is imperative that the quantum nature of electromagnetic radiation is taken into account, and therefore the equations of motion for the electric and magnetic field operators are the Heisenberg equations. However, it can be shown⁹ that these equations are identical in form to the classical Maxwell equations, and therefore the solution for the (quantum) electric field near a dielectric layer carries a great resemblance with the equivalent classical solution. In particular, the plane waves which make up the vacuum electric field in empty space can now be considered as incident waves on the dielectric layer.¹⁰ The laws of ordinary reflection and refraction then determine how this vacuum field is reflected at the boundaries and how it is partially transmitted through the layer. The amplitude, including the phase, of a reflected and transmitted wave with respect to the amplitude of the incident wave can then be expressed in terms of the Fresnel coefficients R_σ and T_σ , respectively, where $\sigma = s, p$ indicates the polarization of the waves (surface polarized or plane polarized). These Fresnel coefficients are frequency dependent through the frequency dependence of the dielectric constant, and they vary with the angle of incidence. For a given incident wave, the two relevant Fresnel coefficients are related as

$$|T_\sigma|^2 + |R_\sigma|^2 = 1 \quad ,$$

assuming a lossless medium. An explicit expression for the vacuum electric field \underline{E}' near a dielectric layer can now easily be constructed from these considerations. This modified field is then coupled to the atomic dipole according to $H'_I = - \underline{\mu} \cdot \underline{E}'$, which in turn determines the change in the relaxation constant A_g , as compared to its value in empty space.

The very same procedure can be followed for any configuration of boundaries and any kind of material, and in particular for a transparent four-wave mixing PC. The major differences between a DL and a PC are the following: (i) The reflected wave at a PC counterpropagates the incident wave, whereas for a DL this wave travels into the specular direction. It is easy to show^{11,12} that this phase-conjugated wave has as its spatial part (its phase) the complex conjugate of the spatial part of the incident wave, which explains the name of this device. (ii) In a quantum approach this complex conjugation turns into a Hermitian conjugation, which implies that the annihilation part of \underline{E} is reflected by the PC as a creation-operator field, and vice versa. For a linear medium the creation and annihilation parts of the field never couple. (iii) During the four-wave mixing process, energy can be extracted from the pump beams, which opens the possibility for a nonzero value of A_g . It also implies that the reflected vacuum waves can contain energy which can be detected as real photons.¹³ (iv) The Fresnel coefficients P_σ for reflection of the phase-conjugated waves have a geometrical frequency dependence, which is sharply peaked at the pump frequency.¹⁴ The relation between P_σ and T_σ is now found to be

$$|T_\sigma|^2 - |P_\sigma|^2 = 1 \quad ,$$

which implies $|T_o|^2 \geq 1$. Waves which enter the medium from one side and leave the medium at the other side are always amplified. For the vacuum waves this leads to a so-called 'amplified vacuum'.

It is possible to derive expressions for the relaxation constants A_e and A_g without any further knowledge of the Fresnel coefficients. This is due to the symmetry of the vacuum for rotations about an axis perpendicular to the surface, and for reflections in a plane through that axis.¹⁵ It turns out that there are two distinct relaxation constants associated with a single transition. Depending on whether the matrix element $\langle e|\underline{\mu}|g\rangle$ of the atomic dipole is perpendicular or parallel to the surface of the medium, the relaxation constant assumes one of these two values, whereas for an arbitrary orientation of $\langle e|\underline{\mu}|g\rangle$ the relaxation constant is a linear combination of these two values. With the abbreviation

$$a_\alpha = \frac{\omega_o^3}{3\pi\epsilon_o\hbar c^3} |\langle e|\underline{\mu}_\alpha|g\rangle|^2, \quad \alpha = \perp, \parallel,$$

where ω_o is the atomic transition frequency, the relaxation constants can be parameterized as

$$A_e = \begin{cases} a_\alpha(1 + x_\alpha) & , \quad \text{DL} \\ a_\alpha(1 + y_\alpha) & , \quad \text{PC} \end{cases}$$

$$A_g = \begin{cases} 0 & , \quad \text{DL} \\ a_\alpha y_\alpha & , \quad \text{PC} \end{cases}$$

for the various situations under consideration. The four dimensionless auxiliary functions are given by

$$x_\perp = \frac{3}{2} \operatorname{Re} \int_0^1 du e^{2i\omega_0 hu/c} (1-u^2) R_p \quad ,$$

$$x_\parallel = \frac{3}{4} \operatorname{Re} \int_0^1 du e^{2i\omega_0 hu/c} (R_s - u^2 R_p) \quad ,$$

$$y_\perp = \frac{3}{4} \int_0^1 du (1-u^2) |P_p|^2 \quad ,$$

$$y_\parallel = \frac{3}{8} \int_0^1 du (|P_s|^2 + u^2 |P_p|^2) \quad ,$$

where the integration variable u signifies the cosine of the angle of incidence of a plane wave, and the Fresnel coefficients R_σ and P_σ have to be evaluated for this particular angle. In general, the Fresnel coefficients T_σ will also appear in the expressions for x_α and y_α , but for the present cases they could be eliminated by means of the relations given earlier.

The four functions x_\perp , x_\parallel , y_\perp and y_\parallel account for the deviation of the relaxation constants A_e and A_g from their empty-space values, since in the limit $R_\sigma \rightarrow 0$, $P_\sigma \rightarrow 0$ these four functions vanish. The parameter functions x_\perp and x_\parallel for a dielectric depend on the atom-surface distance h , which results in an oscillatory behavior of A_e as a function of h , as shown in Fig. 1. It

is remarkable that the functions y_{\perp} and y_{\parallel} for a phase-conjugating medium do not depend on h . Also notice that the functions x_{\perp} and x_{\parallel} are determined by R_{σ} , which includes the change of phase at reflection, whereas y_{\perp} and y_{\parallel} only depend on the relative intensity, $|P_{\sigma}|^2$, of the reflected waves. Furthermore, the values of y_{\perp} and y_{\parallel} are always non-negative, which means that the decay rate near a PC is always larger than for an atom in empty space. In contrast, x_{\perp} and x_{\parallel} can either be positive or negative corresponding to an enhancement or inhibition of the decay rate due to the presence of the interface, respectively.

The most striking feature of the general form of the relaxation constants is that A_g acquires a nonzero value for the situation where the medium is a four-wave mixer. This implies that spontaneous transitions from the ground state to the excited state occur when the atom is sufficiently close to the PC. In addition, there must be a production of fluorescence radiation since after the spontaneous excitation the atom can decay to its ground state, a process which is accompanied by the emission of an observable photon. The physical mechanism for this phenomenon is an effective three-photon process at the site of the atom, rather than in the medium, as shown by the energy-conserving diagram in Fig. 2. From the interpretation given in Fig. 2 it follows that the fluorescent photon must have a frequency $2\bar{\omega} - \omega_0$, with $\bar{\omega}$ the pump-laser frequency. Since ordinary fluorescent photons which are emitted during a decay from the excited state to the ground state have a frequency ω_0 , it should be experimentally feasible to observe the occurrence of a three-photon event by measuring the spectral distribution of the spontaneously-emitted radiation. An observation of a spectral emission line at frequency $2\bar{\omega} - \omega_0$ would confirm the existence of spontaneous excitation through three-photon processes, as depicted in Fig. 2.

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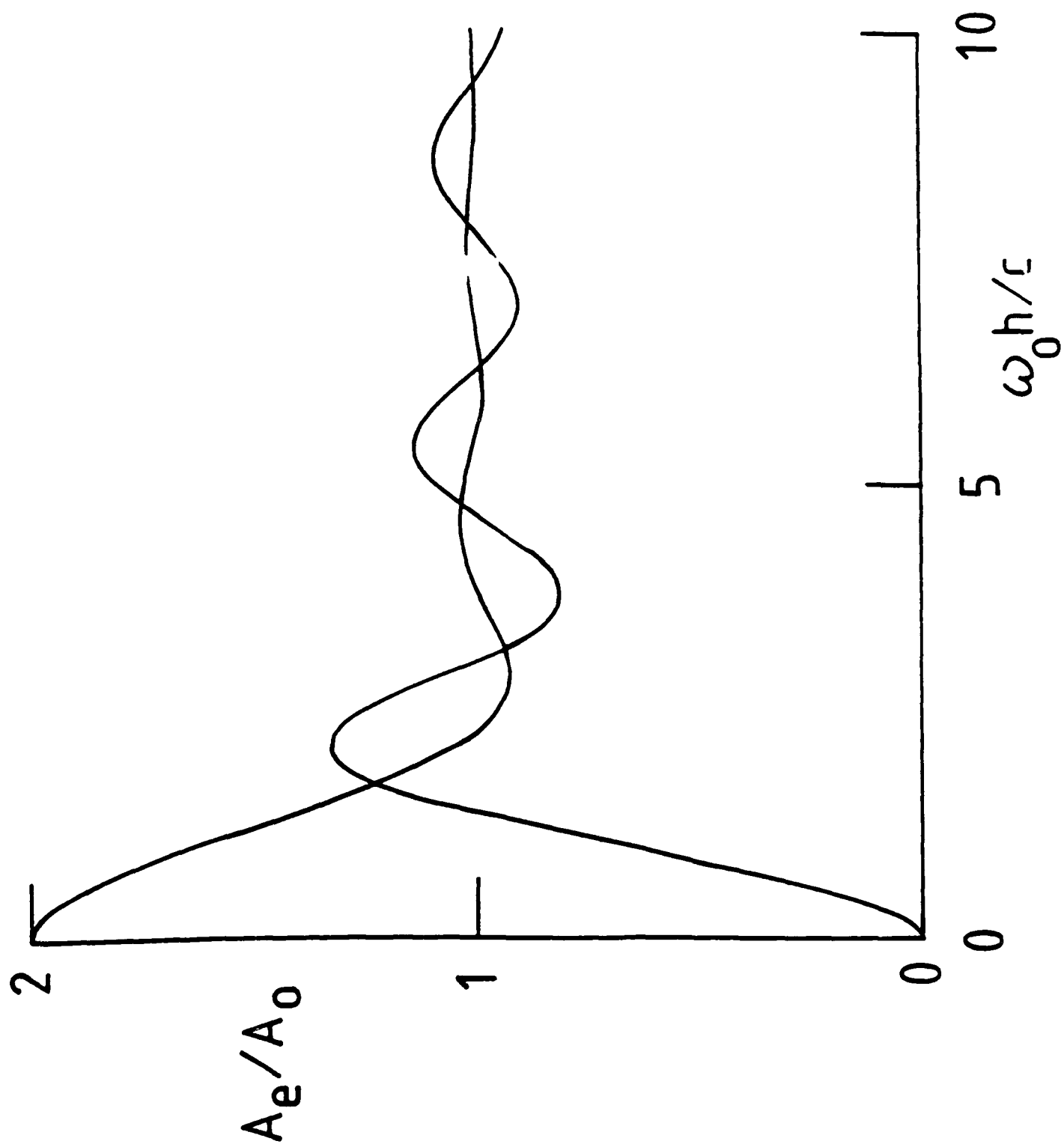
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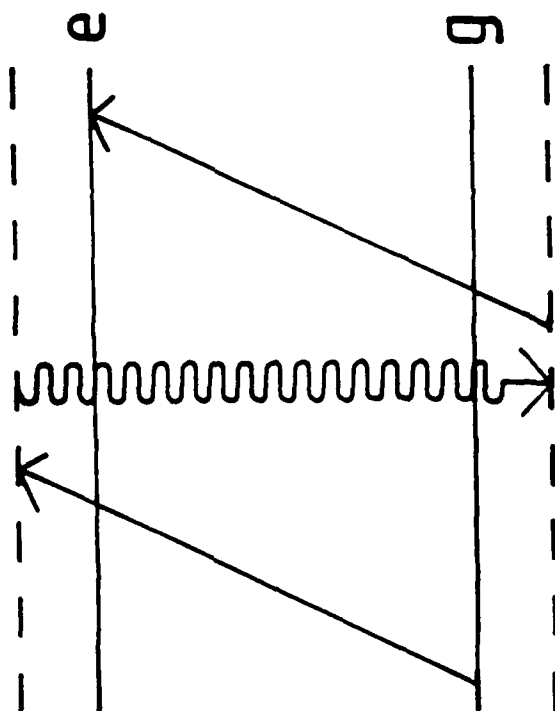
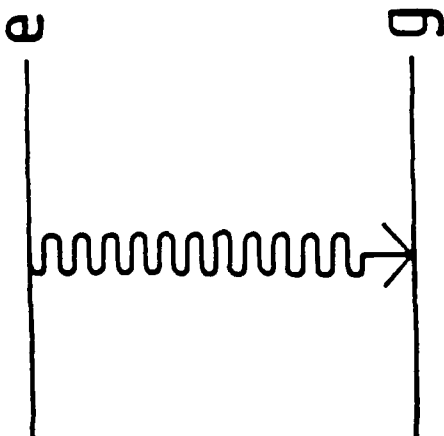
FIGURE CAPTIONS

Fig. 1. Plot of the relaxation constant A_e for a perfect conductor in units of its value A_0 for decay in empty space, versus the normalized atom-surface separation h . The curve with $A_e = 2A_0$ for $h = 0$ pertains to a dipole direction perpendicular to the surface, whereas the second curve gives the value of A_e/A_0 for an atomic dipole moment which is directed parallel to the surface of the mirror. For other dipole directions the value of the relaxation constant is in between the two curves. Notice that for large h the value of A_e approaches A_0 , as it should.

Fig. 2. Illustration of the three-photon relaxation process for an atom near a PC, which gives rise to the nonzero value of A_g (left diagram). The atom is initially in its ground state. The left straight arrow indicates an absorption of a photon with a frequency equal to the frequency $\bar{\omega}$ of the lasers which pump the nonlinear crystal. A subsequent emission of a fluorescent photon (wiggly arrow) brings the atom back to the lower state, and finally a second photon with frequency $\bar{\omega}$ is absorbed, leaving the atom in its excited state. It follows from the diagram that the fluorescent photon must have a frequency $2\bar{\omega} - \omega_0$, which makes it distinguishable from an ordinary fluorescent photon with frequency ω_0 . After this three-photon event the atom can decay to the ground state under emission of an ω_0 -photon (diagram on the right). Both diagrams together form a complete cycle, where the atom starts and ends in the ground state, and this can be considered as a four-wave mixing process at the site of the atom, rather than in the medium. Effectively, the atom extracts two photons with frequency $\bar{\omega}$ from the medium, and converts them into two observable fluorescent photons with frequencies $2\bar{\omega} - \omega_0$ and ω_0 , respectively.

Fig. 1





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